

**PREPARATION AND VALIDATION OF GROSS ALPHA/BETA
SAMPLES USED IN EML'S QUALITY ASSESSMENT PROGRAM**

Salvatore C. Scarpitta

**Environmental Measurements Laboratory
U.S. Department of Energy
201 Varick Street, 5th Floor
New York, NY 10014-4811**

October 1997

19980903 075

DISCLAIMER

"This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof."

This report has been reproduced directly from the best available copy.

Available to DOE and DOE Contractors from the Office of Scientific and Technical Information, P. O. Box 62, Oak Ridge, TN 37831; prices available from (423) 576-8401.

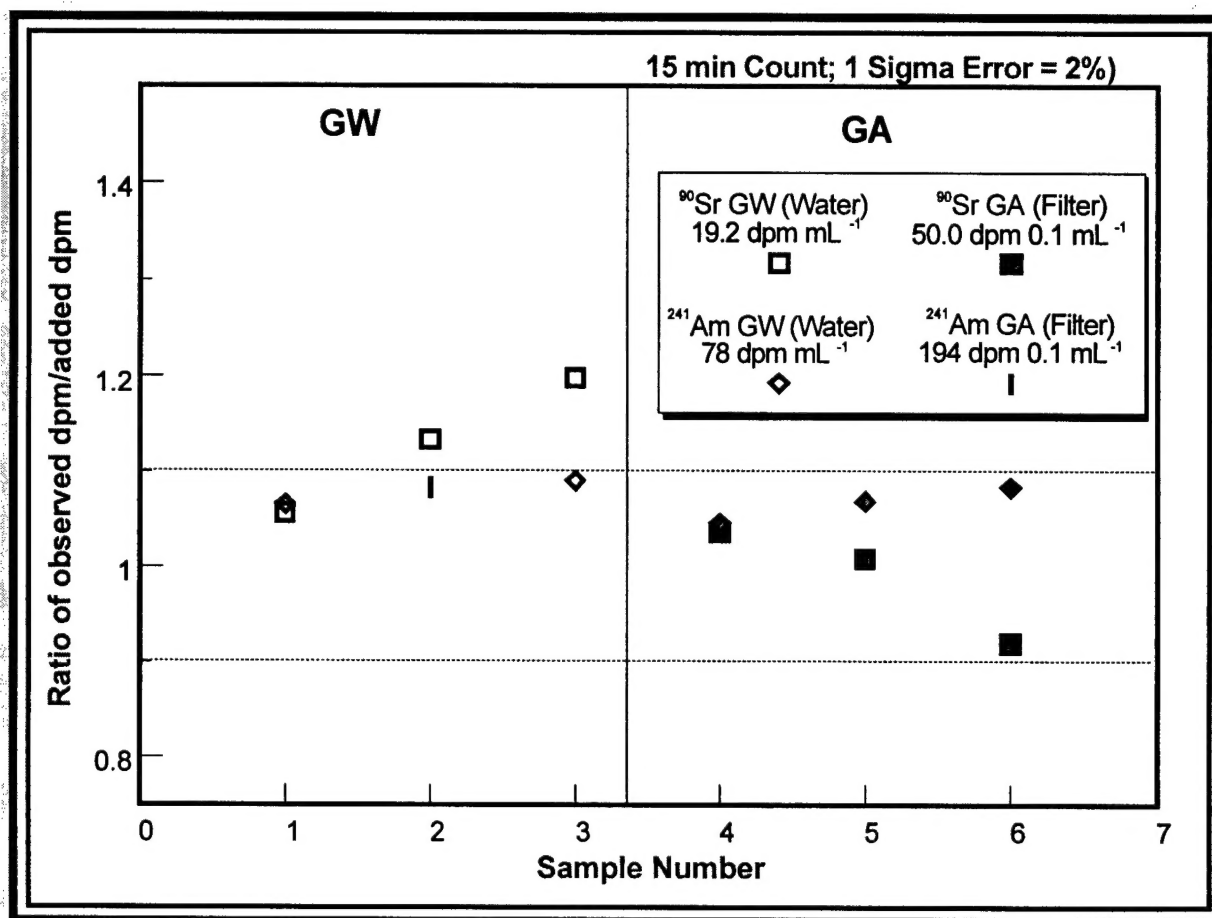
Available to the public from the U.S. Department of Commerce, Technology Administration, National Technical Information Service, 5285 Port Royal Road, Springfield, Virginia 22161, (703) 487-4650.

DTIC QUALITY INSPECTED 1

AQ I 98-11-2339

ENVIRONMENTAL MEASUREMENTS LABORATORY

Preparation and Validation of Gross Alpha/Beta Samples Used in EML's Quality Assessment Program



Salvatore C. Scarpitta

October 1997

**PREPARATION AND VALIDATION OF GROSS ALPHA/BETA
SAMPLES USED IN EML'S QUALITY ASSESSMENT PROGRAM**

Salvatore C. Scarpitta

**Environmental Measurements Laboratory
U.S. Department of Energy
201 Varick Street, 5th Floor
New York, NY 10014-4811**

October 1997

DISCLAIMER

"This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof."

This report has been reproduced directly from the best available copy.

Available to DOE and DOE Contractors from the Office of Scientific and Technical Information, P. O. Box 62, Oak Ridge, TN 37831; prices available from (423) 576-8401.

Available to the public from the U.S. Department of Commerce, Technology Administration, National Technical Information Service, 5285 Port Royal Road, Springfield, Virginia 22161, (703) 487-4650.

A

BSTRACT

A set of water and filter samples have been incorporated into the existing Environmental Measurements Laboratory's (EML) Quality Assessment Program (QAP) for gross alpha/beta determinations by participating DOE laboratories. The participating laboratories are evaluated by comparing their results with the "EML value." The preferred EML method for measuring water and filter samples, described in this report, uses gas flow proportional counters with 2 in. detectors. Procedures for sample preparation, quality control and instrument calibration are presented.

Liquid scintillation (LS) counting is an alternative technique that is suitable for quantifying both the alpha (^{241}Am , ^{230}Th and ^{238}Pu) and beta ($^{90}\text{Sr}/^{90}\text{Y}$) activity concentrations in the solutions used to prepare the QAP water and air filter samples. Three LS counting techniques (Cerenkov, dual dpm and full spectrum analysis) are compared. These techniques may be used to validate the activity concentrations of each component in the alpha/beta solution before the QAP samples are actually prepared.

T

ABLE OF CONTENTS

Abstract	i
Introduction	1
Counting Methods	1
Instrumentation	2
Gas Flow Proportional Counters	2
Efficiency Calibration and Reference Standards for	
Gas Flow Proportional Counters	3
Calibration	3
Cross talk and Self-absorption	4
Quality Control (QC)	5
Inter-laboratory QC	5
Inter-laboratory QC	5
Material Preparation	6
Water and Filter Preparation	7
Summary of Procedure	7
Water	7
Filters	8
Step-by-Step Preparation of QAP Gross Alpha/Beta Filters	8
Data Validation	10
Validation of Gross Alpha/Beta Master Solutions by LS Counting	10
Filter Validation	10
EML LS Validation Procedure (for Master Solutions)	10
Special Apparatus	12
Reagents	12
References	13
Table 1	14
Figures 1-10	15-24
Appendix I	25

I NTRODUCTION

Total alpha/beta measurements are done routinely within the U. S. Department of Energy's (DOE)/Environmental Management (EM) complex as a rapid method to estimate the activities of alpha and beta radionuclides in water, air filters, soils, sludge, and waste water. Gross screening analyses are intended to provide the rapid information associated with a particular action level, with a minimum amount of chemical preparation and chemical waste.

The purpose of gross alpha and beta measurements is to: (1) provide adequate information concerning the activity within samples, and, thus, to determine if further detailed analyses are required, (2) support accountability of radioactive materials and ensure that a receiving laboratory's radioactive materials license or limits are not exceeded, and (3) ensure that the Department of Transportation (DOT) regulations concerning the transport of radioactive materials have not been violated.

One component of DOE's Quality Assurance Program is participation in the EML Quality Assessment Program (QAP) by contractor laboratories conducting work for DOE (DOE Order 5400.1, Ch 4.10.C). The EML QAP is a DOE complex-wide effort that provides the improved analytical confidence needed to address environmental restoration and waste management problems and requirements. The 20-year-old EML QAP has been expanded to incorporate additional water and air filter samples for gross alpha/beta analyses, which are distributed concurrently with the routine semiannual QAP samples.

C OUNTING METHODS

Gross alpha/beta measurement techniques are applicable for alpha emitters having energies above 3 MeV and for beta emitters having energies above 0.1 MeV. Both the gas flow proportional counters and the LS counters can be used for these measurements. At EML, the gas flow proportional counters are used to determine the activity concentrations of the water and Styrofoam filter disc samples. The LS counters are used to determine the activity concentrations of both the alpha and beta emitters in the solutions used in the preparation of QAP water and Styrofoam filter disc samples.

Since the radioactive isotopes in the QAP samples are not separated from the solids of the sample, the solid concentration is a limiting factor in determining the sensitivity of the gas flow proportional counting because of self-absorption of alpha and/or beta particles by the sample itself. Water samples are evaporated to a small volume and transferred quantitatively to a stainless steel planchet. The sample residue is dried to a constant weight by heating, and then it is counted in a gas flow proportional counter for alpha and/or beta activity. Because alpha and beta particles are attenuated or scattered by the sample solids, calibration curves

for self-adsorption and scattering must be established if there is mass in a sample. Sample density on the planchet area should not exceed 10 mg cm⁻² for gross alpha, and 20 mg cm⁻² for gross beta for a 2 in. diameter counting planchet. Moisture absorbed by the dried sample residue is an interferant because it enhances self-absorption.

When measuring alpha and beta particle activity using a gas-flow proportional system, counting at the alpha voltage plateau discriminates against beta particle activity, whereas counting at the beta voltage plateau is somewhat sensitive to the alpha particle activity present in the sample. This phenomenon is termed "cross talk" and is compensated for during instrument calibrations. The "cross talk" factors are determined using alpha (^{238,239}Pu or ²⁴¹Am) and beta (⁹⁰Sr or ¹³⁷Cs) solutions of known activity concentrations counted over a range of absorber thicknesses (mg cm⁻²) encountered during routine sampling and counting.

Master solutions and water samples can be counted for gross alpha/beta activity using LS counters. The procedure applies to clear liquid samples and can be completed in 1-2 h once the efficiency curves have been established (Scarpitta and Fisenne 1996).

I NSTRUMENTATION

Gas Flow Proportional Counters

EML has a Tennelec/Nucleus, Model LB4100W windowless gas-flow low-level alpha/beta counting system (Oxford Instrument Co., Nuclear Measurements Group, 601 Oak Ridge Turnpike, TN 37831-2560). The gridded proportional detectors reportedly eliminate "cross talk" between the alpha and beta channels, allowing for the simultaneous detection of gross alpha and gross beta activities. The basic unit consists of four drawers with four 1-in. or 2-in. detectors and one guard detector per drawer. The calibration sources of ⁹⁰Sr ($t_{1/2} = 27.7$ y) and ²¹⁰Po ($t_{1/2} = 138.4$ d) used for both plateau voltage and efficiency calibrations are listed in Table 1. Typical alpha/beta plateau voltages and point source efficiencies for the 2 in. detectors (drawers C and D) are shown in Figures 1 and 2, respectively. The alpha efficiency is typically about 40%, whereas that of ⁹⁰Sr (in equilibrium with its progeny, ⁹⁰Y) is about 60-70%.

All of the data are processed with a 486DX (33 MHz) PC computer that operates in both DOS and Windows. The computer system is compatible with commercially available data analysis spreadsheet software (i.e., Excel, Quatro, Minitab) for Windows.

The 2-in. diameter gas proportional detectors have a manufacturer measured alpha background of 0.05 counts min⁻¹ and a beta background of 1.0 counts min⁻¹. An instrument with a 1.0 counts min⁻¹ beta background will have an LLD of 37 mBq (1 pCi) for a 60-min count at the 95% confidence level, with a 30%

detection efficiency, whereas an instrument with an alpha background of 0.05 counts min⁻¹ will have an LLD of 67 mBq (or 0.2 pCi = 0.4 dpm) for the same count time and detection efficiency (NCRP 1978). This eight-detector system can accommodate a throughput of 56 samples per hour if each sample is counted for 15 min.

The LS system at EML is a multisample, automatic, Packard Tri-Carb 2250CA that can detect both alpha and beta particles. The system can be operated in either the "dual isotope" or "full spectrum analysis" mode to discriminate between alpha and beta particles (EML Procedures Manual 1997, Procedure Ba-01-R).

EFFICIENCY CALIBRATION AND REFERENCE STANDARDS FOR GAS FLOW PROPORTIONAL COUNTERS

Calibration

For absolute gross alpha and gross beta measurements, each detector must be calibrated to obtain the efficiency, which is the ratio of the count rate to the disintegration rate. Alpha emitting ²⁴¹Am will be used initially as a reference standard to be substituted in future QAPs with ²³⁰Th or ²³⁸Pu, each of which is close to the energy of the alpha particles emitted by naturally occurring ²³⁸Th and ²²⁴Ra. Styrofoam standards are prepared in the geometry and weight ranges to be encountered in these gross analyses. Isotopes used for QAP purposes are NIST-traceable (National Institute of Standards and Technology, Radioactivity Group, Building 245, Room C114, Gaithersburg, MA 20899).

⁹⁰Sr/⁹⁰Y and ¹³⁷Cs have been used extensively as the beta component for gross beta activity. Standard solutions of each of these radionuclides are readily available. However, cesium is volatile at elevated temperatures (above 450°C). Some water supplies have dissolved solids (salts) that, when converted to nitrate salts, are quite hygroscopic and need to be converted to oxides by heating to a red heat to obtain sample aliquots that are weight-stable. Sample weight stability is essential to gross alpha and gross beta measurements to ensure the accuracy of the self-absorption counting efficiency factor to be used for the samples. ⁹⁰Sr in equilibrium with its daughter ⁹⁰Y is the preferred radionuclide for gross beta calibrations although beta emitting ⁹⁹Tc, with a much longer half-life, may be used. The disadvantage is that technetium is volatile at elevated temperatures.

Cross Talk and Self-absorption

Initially, QAP water and simulated filters will be prepared using matrix-free solutions. When and if salts are added to the master solutions, then self-absorption calibration curves must be generated. For each gas

proportional detector, separate alpha and beta particle self-absorption graphs would be established that showed the water sample residue weight (mg) vs. the efficiency factor (counts min⁻¹/dpm), using standard alpha and beta emitter solutions and deionized water containing varying amounts of added salts. For the alpha calibration curve, alpha activity is added to varying sizes of aliquots of water containing a fixed amount of salts such that the aliquot residue weight is varied between 0 and 100 mg (for a 2-in. counting planchet). A similar calibration curve would be prepared with standard beta activity and salted water aliquots, varying the residue weight between 0 and 300 mg (for a 2 in. planchet). If it is planned to use water-sample aliquot volumes that always contain 100 mg of dried water solids, then only the efficiency factor for that residue weight needs to be established.

Water aliquots, with an added NIST traceable ²⁴¹Am (3 Bq) or ⁹⁰Sr (1 Bq) standard, are acidified with a few milliliters of 16N HNO₃ and evaporated onto 1-7/8 in. Styrofoam discs. The discs are dried at 105°C for 15 min under a heat lamp before counting. Care is taken not to allow the disks to deform. Three to five minute counts are obtained on each disc and in each drawer to determine detector efficiencies and "cross talk" factors. Figures 3 and 4 show the detector efficiencies, background (using blank foam pads) and "cross talk" factors for the 2 in. detectors in drawers C and D of the Oxford/Tennelec LB4100 System.

QUALITY CONTROL (QC)

The QC aspect of EML's gross alpha/beta QAP samples involves both intra-laboratory and inter-laboratory QC. Intra-laboratory QC is: (1) procedure development, instrument maintenance, record keeping, and (2) QAP sample preparation, gross alpha /beta determination and data validation before distribution. Inter-laboratory QC involves participation in an external quality assurance monitoring program.

Intra-laboratory QC

EML will participate in the U. S. Environmental Protection Agency (EPA) Performance Evaluation Studies (PES) Program, which is an agency-wide quality assurance program assigned to the Nuclear Radiation Assessment Division of the EPA at the Environmental Monitoring Systems Laboratory, Quality Assurance Branch (EMSL-LV, P.O. Box 15027, Las Vegas, NV 89114.). Participation in the PES program will be useful for documenting precision and accuracy, and for helping to indicate instrumental or procedural problems. Participation in the PES program will also be useful in augmenting the EML QC program by serving as a check on EML's internal QC program.

EPA typically provides 1 L samples containing a mixture of radionuclides preserved with 0.5N HCl. These samples are distributed semiannually and consist of two samples per shipment. Sample (A) contains ^{226}Ra , ^{228}Ra and natural U, which are analyzed for gross alpha activity. Sample (B) contains ^{89}Sr , ^{90}Sr , ^{60}Co , ^{134}Ce and ^{137}Cs ($< 7.4 \text{ Bq L}^{-1}$), which are analyzed for gross beta activity. Three 2 in. diameter Styrofoam discs containing ^{241}Am , ^{90}Sr or ^{137}Cs ($< 7.4 \text{ Bq filter}^{-1}$) are distributed annually by EPA. These discs will be analyzed at EML for gross alpha and beta activities concurrently with the QAP gross alpha/beta water and air filter samples.

Inter-laboratory QC

All EML QC data, including background data and calibration standards, are maintained as part of the software package of the Oxford alpha/beta detection system. The data will be available for easy reference or inspection and for production of QC charts. During gross alpha/beta air filter and water sample counting, blanks will be interspersed between the actual samples to determine if detector contamination is occurring. Other QC measures are outlined later in this report. Previously measured calibration samples and/or standard reference materials (SRM) will be routinely used to ensure that correct procedures are being followed and that all equipment is operating properly.

MATERIAL PREPARATION

Samples for gross alpha and beta counting that are distributed to DOE/QAP participating laboratories consist of artificially spiked water and Styrofoam filter disc samples. Distilled or deionized water (Type II) having a resistance value between 0.5 and 2.0 megohms (2.0 to 0.5 mhos) cm^{-1} at 25°C will be used. Master solutions of known activity will be prepared using NIST traceable radionuclides calibrated at EML using a 2π geometry detecting system (EML Procedures Manual 1997, Procedure A-01-R). The master solutions are acidified with 1N HCl to prevent adsorptive losses to the walls of the mixing vessels.

The final 1 L spiked water master solution contains $< 7.4 \text{ Bq mL}^{-1}$ of an alpha/beta mix of ^{90}Sr , and $^{238,239}\text{Pu}$, ^{241}Am , or natural U, yielding 250 4-mL samples. The gross alpha/beta water samples are dispensed into 4 mL precleaned glass vials with leak-proof Teflon sealed caps for the semiannual QAP distribution.

The water samples for gross alpha/beta measurements are typically carrier free, unless salts are added to the master tracer solution at the time of preparation. A 4-mL aliquot of a water sample would contain no more than 100 mg mL^{-1} (for alpha only or alpha and beta determination) or 200 mg mL^{-1} (for beta only determination) of total water solids.

Each participating laboratory is supplied with sufficient alpha and beta emitting activity ($< 1.7 \text{ Bq mL}^{-1}$) to perform the measurements from water. The water sample contains only one alpha and one beta NIST traceable radionuclide (i.e., ^{241}Am and ^{90}Sr). An aliquot of the sample can be transferred quantitatively and evaporated onto a 1 in. or 2 in. metal planchet before alpha and beta measurements as described in the next section, Filter Preparation. Spiked Styrofoam filter discs of a 1-7/8 in. diameter are prepared at EML in the manner described later in this report. Each filter is individually counted for gross alpha/beta activity to establish sample homogeneity, determine outliers, and establish the EML measured value as described in the next section, Filter Preparation.

WATER AND FILTER PREPARATION

Summary of Procedure

An aliquot of an acidified water sample is evaporated to a small volume and transferred quantitatively to a tared 1 in. or 2 in. stainless-steel counting planchet. The sample residue is dried to a constant weight, reweighed to determine dry residue weight, and then counted for alpha and/or beta radioactivity. Styrofoam filter discs are spiked with 240 μL of spiking solution, dried, then directly counted, without chemical processing. Counting efficiencies for both alpha and beta particle activities are selected according to the amount of sample solids from counting efficiency vs. sample solids standard curves. The same calibration standards are used to determine the "cross talk" factors.

Water

The water analysis method presented here is identical to other standard methods (DOE RP710, EPA method 9310). If the water sample contains hygroscopic salts, the planchette samples must be heated first at a low temperature and then flamed to a dull red to convert the salts to their respective oxide forms. Both filter and water samples may be covered with a 0.00025-in. thick Mylar film. The efficiency and cross talk curves generated from calibration standards must be based on the same sample configuration.

Evaporate the aliquot in a beaker to near dryness on a hot plate. If water samples are known or suspected to contain chloride salts, those chloride salts should be converted to nitrate salts before the sample residue is transferred to a stainless-steel planchet (chlorides will attack stainless steel and increase the sample solids, and no correction can be made for those added solids). Chloride salts can be converted to nitrate salts by adding 5-mL portions of 16N HNO_3 to the sample residue and evaporating to near dryness (two treatments are usually sufficient). Add 2 mL of 0.5 N HNO_3 to the beaker and swirl to dissolve the residue. Quantitatively transfer the aliquot concentrate in small portions (not more than 1 mL at a time) to a tared planchet, evaporating each portion to dryness. Dry the sample residue in a drying oven at 105°C for at least 1 h, cool in a desiccator, weigh and count. Store the sample residue in a desiccator until ready for counting.

Some types of water-dissolved solids, when converted to nitrate salts, are quite hygroscopic even after being dried at 105°C for 1 h. When such hygroscopic salts are present in samples that are put into an automatic counting system, those samples gain weight while they are waiting to be counted, and will result in inaccurate counting data. When there is evidence of hygroscopic salts in sample counting planchets, it is recommended that they be flamed to a dull red heat with a Meeker burner for a few minutes to convert the nitrate salts to oxides before weighing and counting. (Having a loss of cesium or technetium during the flaming of the samples is possible.)

Count for alpha and beta activity at their respective voltage plateaus if the detection unit does not perform the measurement automatically. If the sample is to be recounted for reverification, store it in a desiccator. The calculations are done manually as described in Appendix I of this report or automatically by the computer software package.

Filters

Approximately 150 to 200 1-7/8 in. diameter spiked Styrofoam filter discs are prepared biannually for the QAP gross alpha/beta measurements. The actual spiking of the filter discs is performed using a mechanical automated pipetter developed at EML that is capable of delivering predetermined microliter amounts of tracer solution onto the filter surface. The pipetting unit can deliver 12 aliquots of 20 μL each onto a 2 in. diameter disc in a fixed geometric pattern and it is accurate to within 8% of the preset amount.

Each spiked filter is dried under a heat lamp ($<100^{\circ}\text{C}$) and stored in a prenumbered plastic sample holder. The analysis of the filter discs requires no prechemistry or sample preparation. After each filter is counted, the data is downloaded to an appropriate spreadsheet (Minitab, Excel or Quatro Pro) for statistical testing of the data. This is done to eliminate outliers and to determine the mean EML measured value.

Each filter is finally packaged in a "zip-lock" plastic bag that may contain a commercially available desiccant pellet to reduce moisture uptake by the sample during transport.

STEP-BY-STEP PREPARATION OF QAP GROSS ALPHA/BETA FILTERS

1. Obtain 45-mm precut Styrofoam filter discs (Hefty Strong Soak Proof Dishes, Mobil Chemical Co., Pittsford, NY 14534).
2. Obtain the QAP gross alpha master solution. Add 1 drop of dye to visualize spots. The solution is acidified with 1N HCl and contains a total activity of $< 3000 \text{ dpm mL}^{-1}$ of alpha and beta emitters. (Note: Wear disposable Latex gloves.)
3. Obtain a template for the 45-mm Styrofoam filter discs. Place the template on the turntable of the automatic EML pipetter, and a blank Styrofoam filter disc on the template.
4. Place a 50-mm rubber O-ring on the surface of the Styrofoam filter disc and adjust the angle of the pipetter tip (**not the radius**) so that all 12 drops of water are dispensed inside the O-ring. Mark the position of the arm for future use.

5. Secure 50-mm plastic Millipore filter holders used to store the dry spotted discs. Replace the water vial with the QAP gross alpha solution and prime the pump. Spot three filters and **discard** them.
6. Begin spotting the Styrofoam filter discs, weighing every 10th disc before and after spotting. Record the data. The pipetter should be set to dispense about 0.100 to $0.120 \text{ g filter}^{-1} \pm 2\%$.
7. Secure a thermometer (0 - 200°C) and adjust the height of the heat lamps so that any point on the drying surface does not exceed 105°C . (**Note:** The Styrofoam deforms at 120 - 130°C .)
8. Prepare the drying surface with clean disposable pads. Dry the samples for 15 min on a 45-mm template. Line each slot with glass fiber filter paper to prevent deformation of discs during heating. (**Note:** If a disc deforms, discard it.)
9. Transfer dry filters (spotted side up) to prenumbered plastic Millipore 50 mm holders and secure cap. Use Teflon forceps prewashed in ethanol. Return the filter holders to the rack. Each filter will be individually counted later.
10. Using the worksheet (see Figure 5), obtain the average weight of the gross alpha solution dispensed per filter, 1 sigma error and % coefficient of variation (CV). (**Note:** The data set is required for QC purposes.)
11. Clean the pump of the automatic pipetter with deionized water when the task is completed. Store the QAP gross alpha solution and the 45-mm plexiglass template for future use.

DATA VALIDATION

Validation of Gross Alpha/Beta Master Solutions by LS Counting

The EML Packard Tri-Carb 2250CA LS counter is used to validate the activity concentrations (alpha and beta) of the master solutions used for preparing QAP gross alpha/beta water and gross alpha filter samples. Typically, $^{90}\text{Y}/\text{Sr}$ and ^{241}Am can be used in activity ratios ranging from 1:1 to about 1:4 (see EML's LS validating procedure below). The activity concentrations in gross alpha/beta water should range from 0.167-1.67 Bq mL⁻¹ of ^{90}Sr and 0.8-8 Bq mL⁻¹ of ^{241}Am , whereas those in the gross alpha filter solution should range from 1.7-8.3 Bq mL⁻¹ of ^{90}Sr and 8.3-33.3 Bq of ^{241}Am . Figures 6-8 show the results obtained using full-spectrum analysis or dual dpm analyses (Packard Users Manual 1994) and Cerenkov counting (Scarpitta and Fisenne 1996), respectively, to verify the activity concentrations of both ^{241}Am and ^{90}Sr in the gross alpha filter and gross alpha/beta water master solutions.

Filter Validation

The entire filter set may be validated by: (1) comparison of the "expected" activity level, based on the exact volume of the master spiking solution dispensed gravimetrically, (2) comparison to aliquots of identical spiked solutions counted separately on metal planchets, (3) comparison to an alternative gross alpha/beta counting technique, (4) comparison to the EPA EMSL-LV air-filter sample (when it becomes available), or (5) inclusion of previous QAP gross alpha/beta Styrofoam filter disc samples.

Figure 9 shows the alpha and beta activities on 1-7/8 in. Styrofoam pads each counted for 15 min in the Tennelec LB4100W system. The expected values were obtained from the average mass of the master gross alpha/beta water solutions applied to every tenth filter. Figure 10 shows the reproducibility of the Tennelec LB4100W system for replicate 15-min counts on 10 individual filter discs.

EML LS Validating Procedure (for Master Solutions)

1. Obtain master solutions for gross alpha/beta water (GW) and gross alpha air filters (GA) acidified with 1N HCl. The solution is colored with one drop of dye after data validation.
2. Dispense 5.0 mL of GW solution gravimetrically into 20-mL glass LS vials (in triplicate).
3. Gravimetrically dispense 0.1 - 0.2 mL of GA solution into 20-mL glass LS vials (in triplicate). Add 5 mL of deionized water.
4. Prepare a blank containing 5 mL of deionized water.

5. Obtain a separate reference standard of $^{90}\text{Sr}/^{90}\text{Y}$ (about 1000 dpm g^{-1} of ^{90}Sr). Weigh known amounts of the reference standard (about 0.2 g) into 20 mL glass scintillation vials. Add 5 mL of deionized water (see **Note 1**).
6. Cerenkov count the reference standard, blank and both gross alpha/beta water and filter samples for 15 min each using Protocol #4 of the Packard Tri-Carb 2250CA LS counter without separating the ^{90}Y . The Cerenkov window should be set at 0-15 keV, although a window of 0-50 keV could be used.
7. Determine the ^{90}Y Cerenkov counting efficiency, Eff^{90}Y (typically 70%) according to the method described in Procedure Sr-04 (EML Procedures Manual 1997). Calculate ^{90}Sr activity for each sample using the ^{90}Y efficiency and the mass of the master solution sample, M_s (g).
8. Add 15 mL of Insta-Gel-XF cocktail to 5 mL samples. Shake for 20 s and wipe the vials clean with an ethanol soaked tissue.
9. Recount the samples for 20 min each using Protocol #4 (full spectrum analysis or dual DPM) to determine both the ^{90}Sr and ^{241}Am activity concentrations simultaneously. The results will be printed as dpm ^{90}Sr and dpm ^{241}Am per sample (see **Note 2**).
10. Examples of the results obtained using this method are shown in Figures 6-8.

Notes:

1. Before counting the samples, run the ^3H , ^{14}C and background samples in an LS counter using the System Normalization Check (SNC) samples provided by the manufacturer. This is done to calibrate the LS system and to provide QC checks on the performance of the instrument.
2. Full spectrum analysis requires that two separately quenched calibration curves be established for $^{90}\text{Sr}/^{90}\text{Y}$ and ^{241}Am before processing the unknown samples. The full spectrum analysis method is described in the Packard Instrument Reference Manual. Five calibration samples containing 210 dpm of $^{90}\text{Sr}/^{90}\text{Y}$ and five samples containing 209 dpm ^{241}Am were prepared in February 1994. These samples can be reused to reestablish the quench curves if necessary. The first sample in the rack should always be background. The appropriate windows, QIP and SIS values are currently resident in the LS system as Protocol #3.

SPECIAL APPARATUS

1. Gas-flow proportional counting system, or scintillation detection system.
2. Stainless-steel counting planchets or molded plastic ring/discs.
3. Electric hot plate.
4. Drying oven.
5. Drying lamp.
6. Glassware.
7. Analytical balance.
8. Meeker burner.
9. Mylar film.

REAGENTS

1. All chemicals should be of reagent-grade or equivalent whenever they are commercially available.
2. Nitric acid, 1N - mix 6.2 mL 16N HNO₃ (concentration) with deionized or distilled water and dilute to 100 mL.
3. NIST traceable calibration standards (alpha and beta emitting).

REFERENCES

EML Procedures Manual, HASL-300, 28th Edition, Volumes I and II

N. A. Chieco (Editor)

Available from: NTIS, Springfield, VA 22161, Product No. PB97-162549; EML's Web Site

www.eml.doe/procman/intro.htm (1970)

National Council on Radiation Protection and Measurement

A Handbook of Radioactivity Measurements Procedures

NCRP Report No. 58, Washington, D.C. 20014 (1978)

Scarpitta, S. C., Fisenne, I. M.

Calibration of a LS Counter for Alpha, Beta and Cerenkov Counting

USDOE Report EML-583 (1996)

Tricarb Liquid Scintillation Analyzer Reference Manual, No. 7001300-Rev-B

Packard Instrument Co., 800 Research Parkway, Meriden CT 06450 (1994)

U. S. Department of Energy

DOE Methods for Evaluating Environmental and Waste Management

DOE/EM Report-0089T (1993)

U. S. Environmental Protection Agency

EPA Test Methods for Evaluating Solid Waste

EPA Report No. SW-846 (1996)

TABLE 1

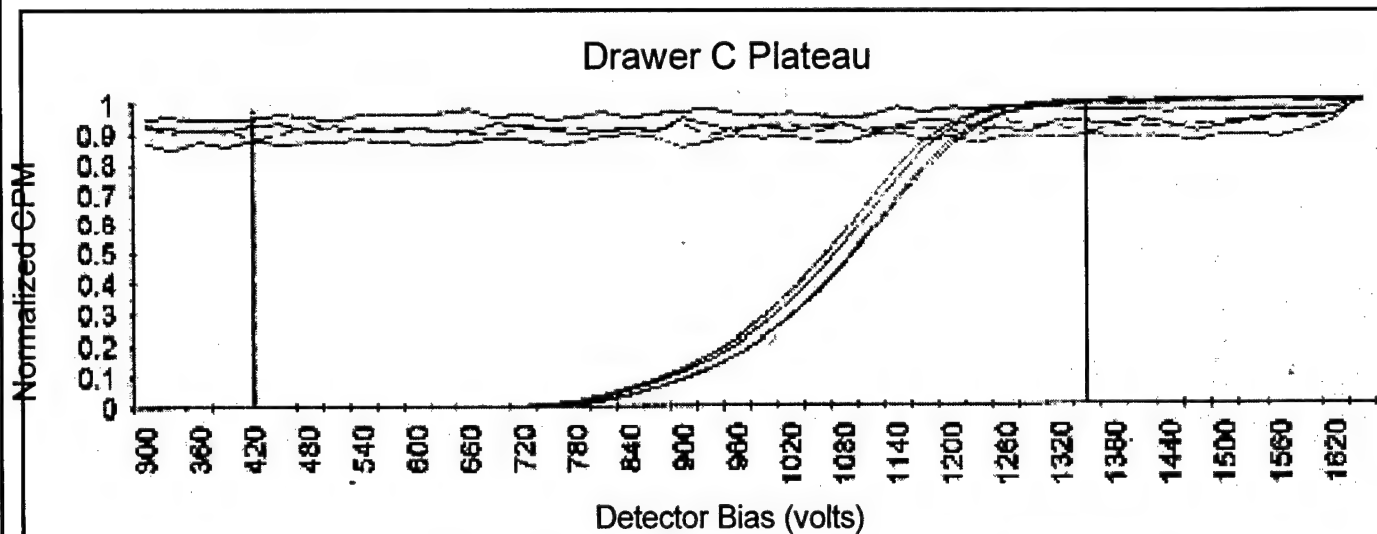
GROSS ALPHA/BETA CALIBRATION POINT SOURCES

Nuclide ID	Activity		
	(nCi)	(dpm)	(Bq)
<hr/> ²¹⁰ Po (01Aug94)*			
B-652	51.26	113,797	1,896.6
B-653	45.31	100,588	1,676.5
B-654	47.89	106,316	1,793.9
B-655	43.66	96,925	1,615.4
 ⁹⁰ Sr (23Jun94)**			
S26-1009-12	19.6	43,512	725.2
S26-1010-07	20.8	46,176	769.6
S26-1009-6	20.0	44,400	740.0
S26-1009-7	21.8	48,396	806.6

* $\lambda = 5 \text{ E-3 d}^{-1}$ ** $\lambda = 6.85 \text{ E-5 d}^{-1}$

Unit Type: LB4100/W
 Date Performed: 1/31/95 14:51
 FileName: PLAT010
 Batch ID: INIT PLAT C&D DRAWERS SO#28036 202.01.95

Unit id: White
 Application Revision: B
 Application Version: Standare

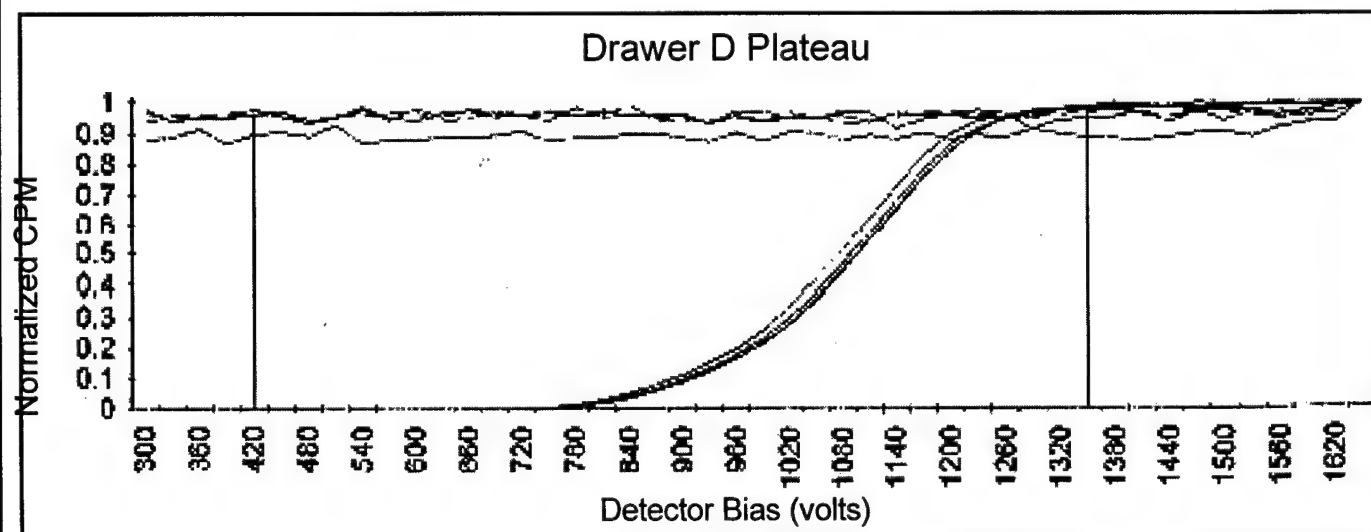


Optimum alpha beta simultaneous operation voltage: 1350
 Optimum alpha only operation voltage: 420

	C1	C2	C3	C4
Betaslop at beta voltage	0.80%	0.08%	0.90%	0.90%

Unit Type: LB4100/W
 Date Performed: 1/31/95 14:51
 FileName: PLAT010
 Batch ID: INIT PLAT C&D DRAWERS SO#28036 202.01.95

Unit id: White
 Application Revision: B
 Application Version: Standare



Optimum alpha beta simultaneous operation voltage: 1350
 Optimum alpha only operation voltage: 420

Figure 1. Voltage plateaus for the Tennelec alpha/beta counters.

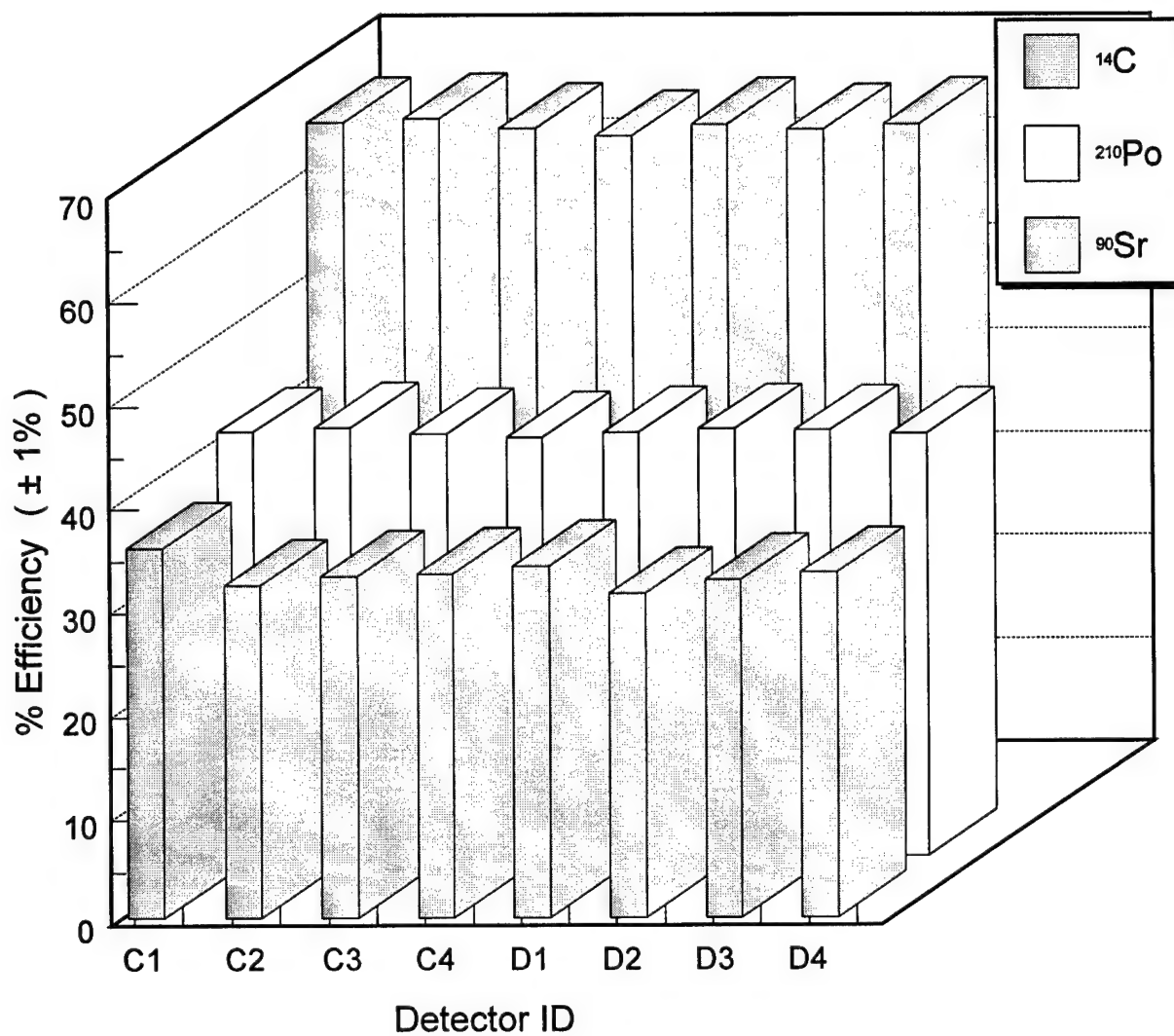


Figure 2. Tennelec gross alpha/beta 2 in. detector efficiencies.

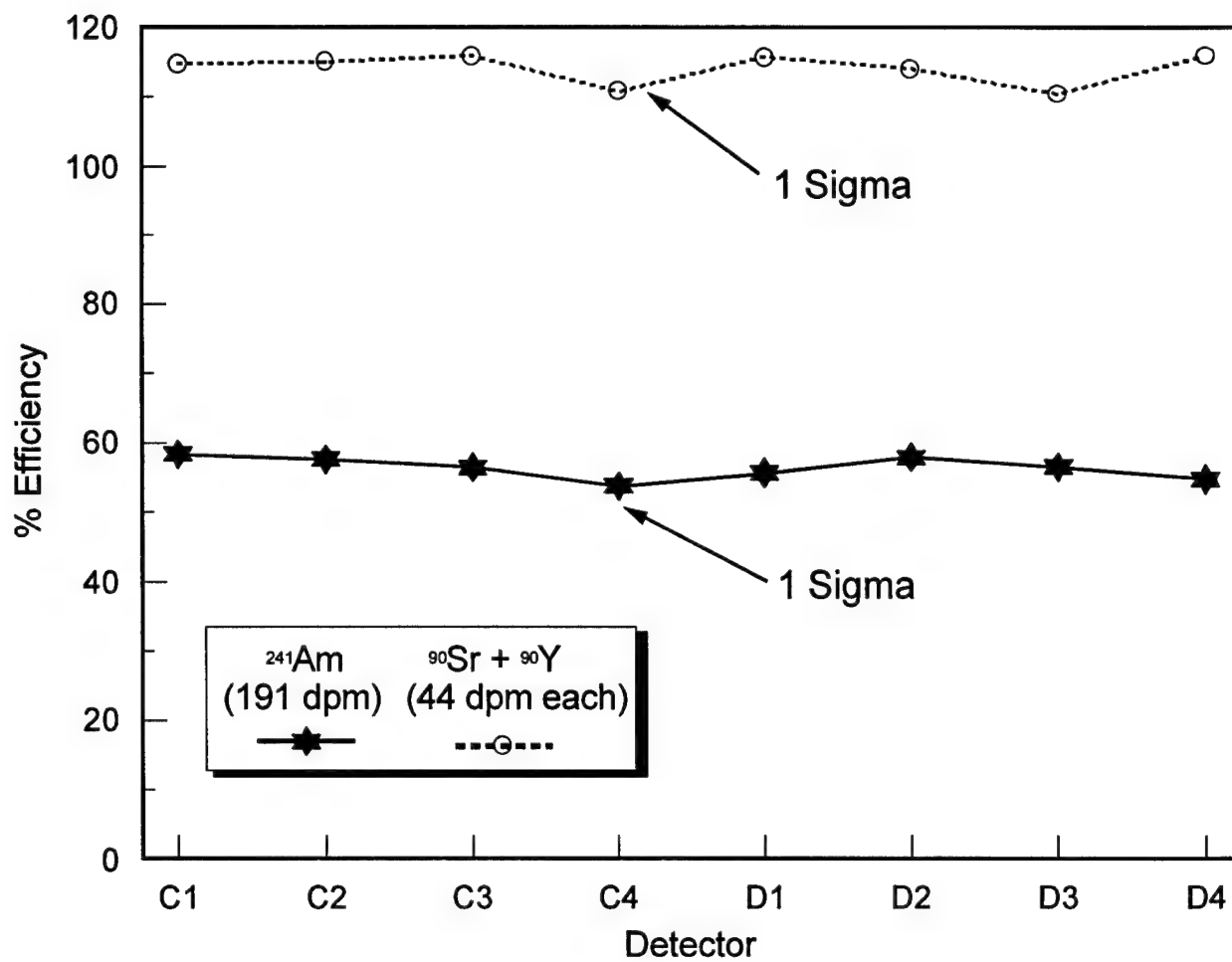


Figure 3. Detector efficiency using 1 7/8 in. foam pads.

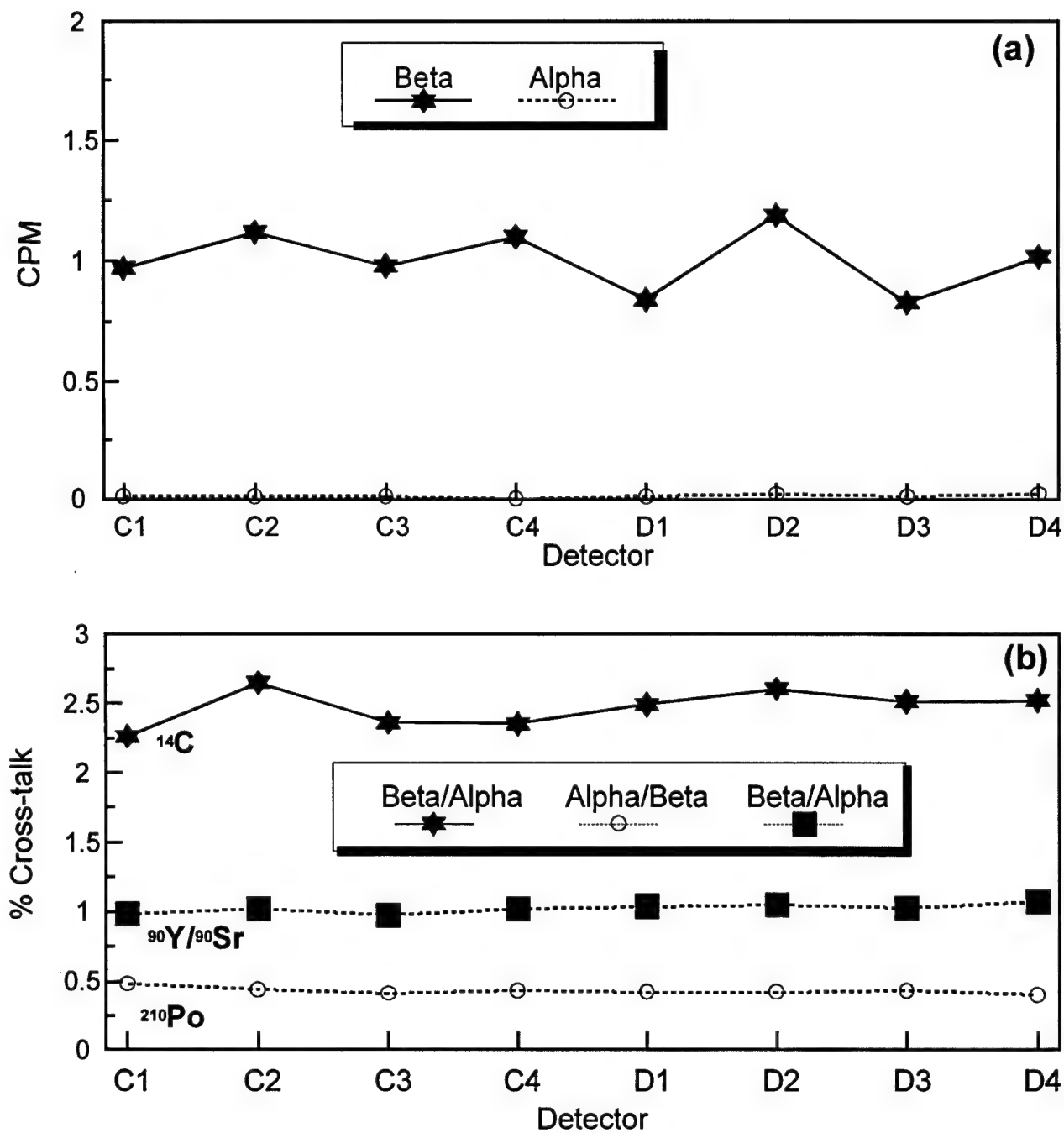


Figure 4. (a) Alpha/beta background for 2 in. detectors; (b) alpha/beta cross-talk for 2 in. detectors..

QAP Gross Alpha and Beta Filters

Weight and Activity Dispensed per Filter

QAP # _____

Filter #

	Tare Weight (gm)	Final Weight (gm)	Weight Dispensed (gm)
10			
20			
30			
40			
50			
60			
70			
80			
90			
100			
110			
120			
130			
140			
150			
160			
170			
180			
190			
200			

Date: _____

Mean Wt. _____

1 Sigma _____

%CV _____

Q.C. Data

_____	dpm ^{90}Sr g $^{-1}$ x Mean Wt. =	_____ dpm ^{90}Sr Filter $^{-1}$
_____	dpm ^{241}Am g $^{-1}$ x Mean Wt. =	_____ dpm ^{241}Am Filter $^{-1}$

Figure 5. QAP gross alpha/beta worksheet.

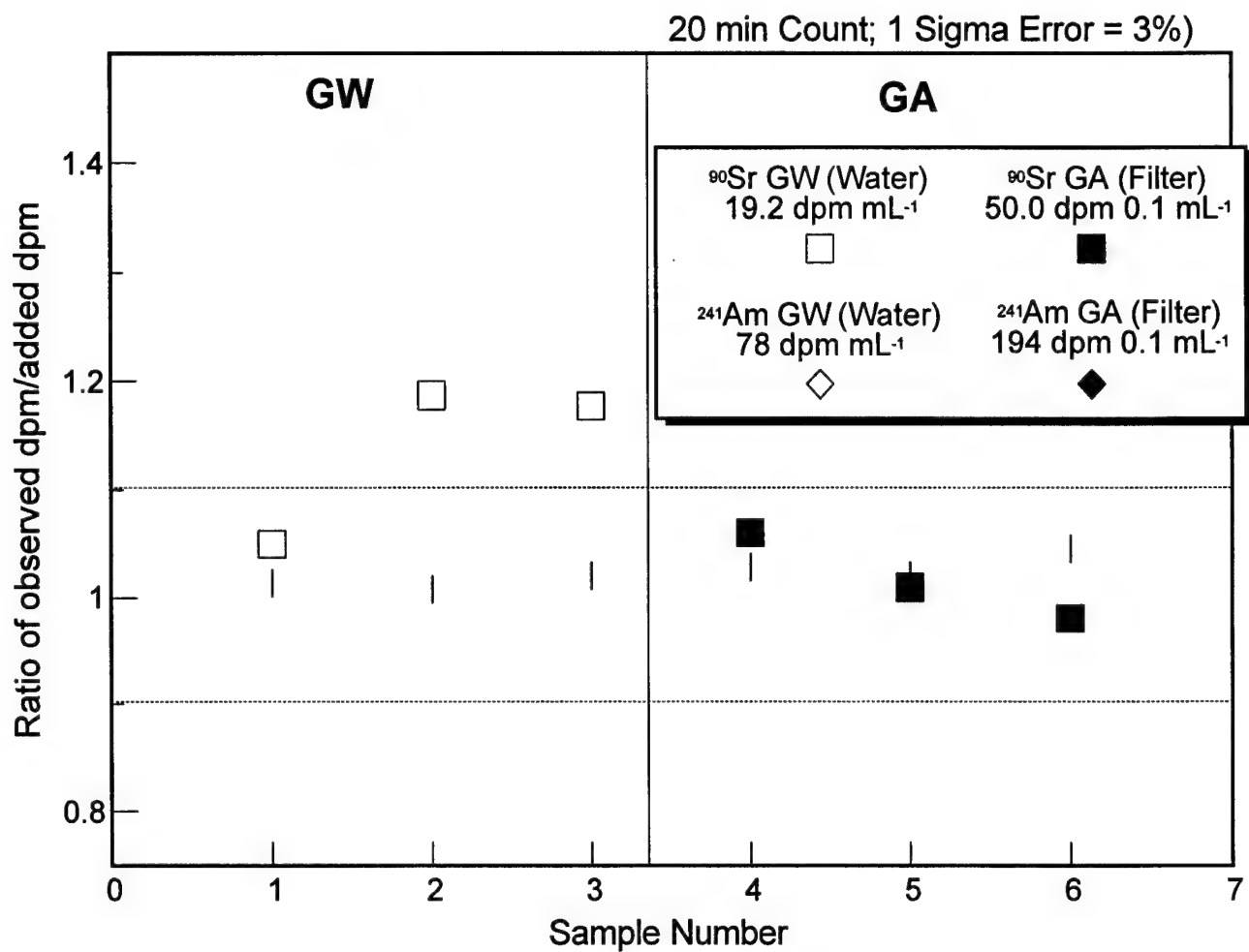


Figure 6. Full spectrum analysis (FSA) results with a Packard Tri-Carb LS counter;.

15 min count; 1 sigma error = 2%

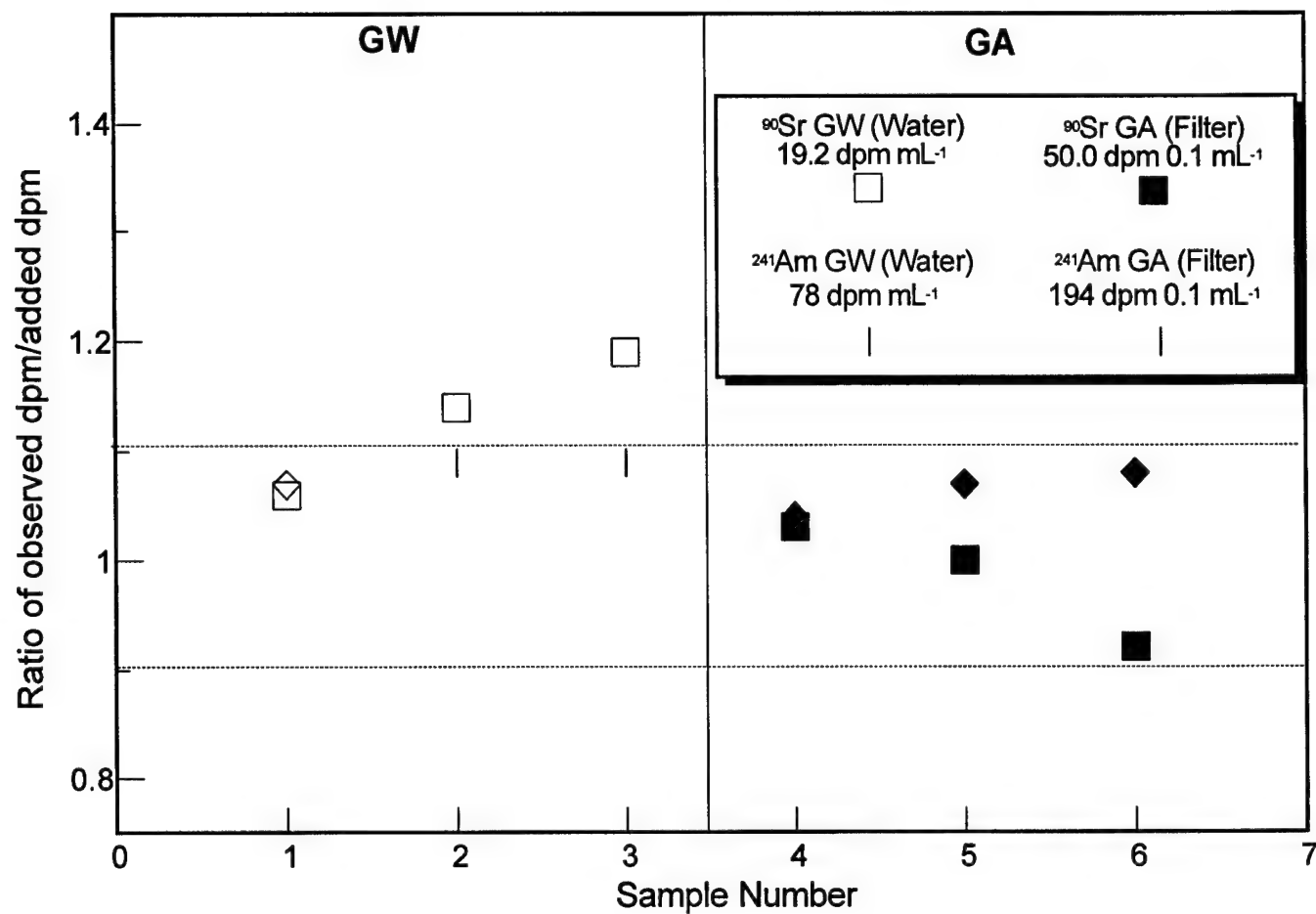


Figure 7. Dual dpm results with a Packard Tri-Carb counter.

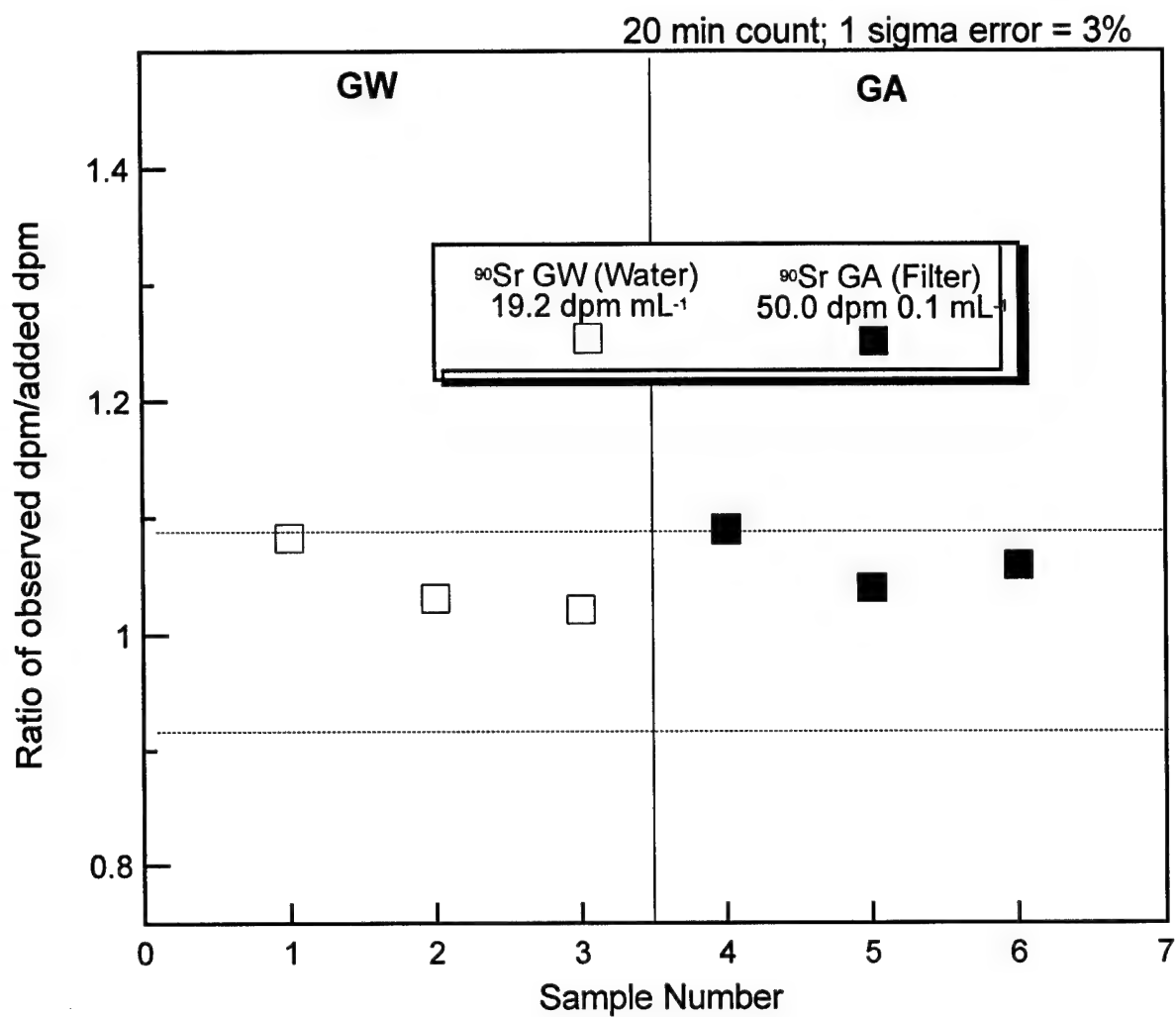


Figure 8. Cerenkov counting results with a Packard Tri-Carb counter.

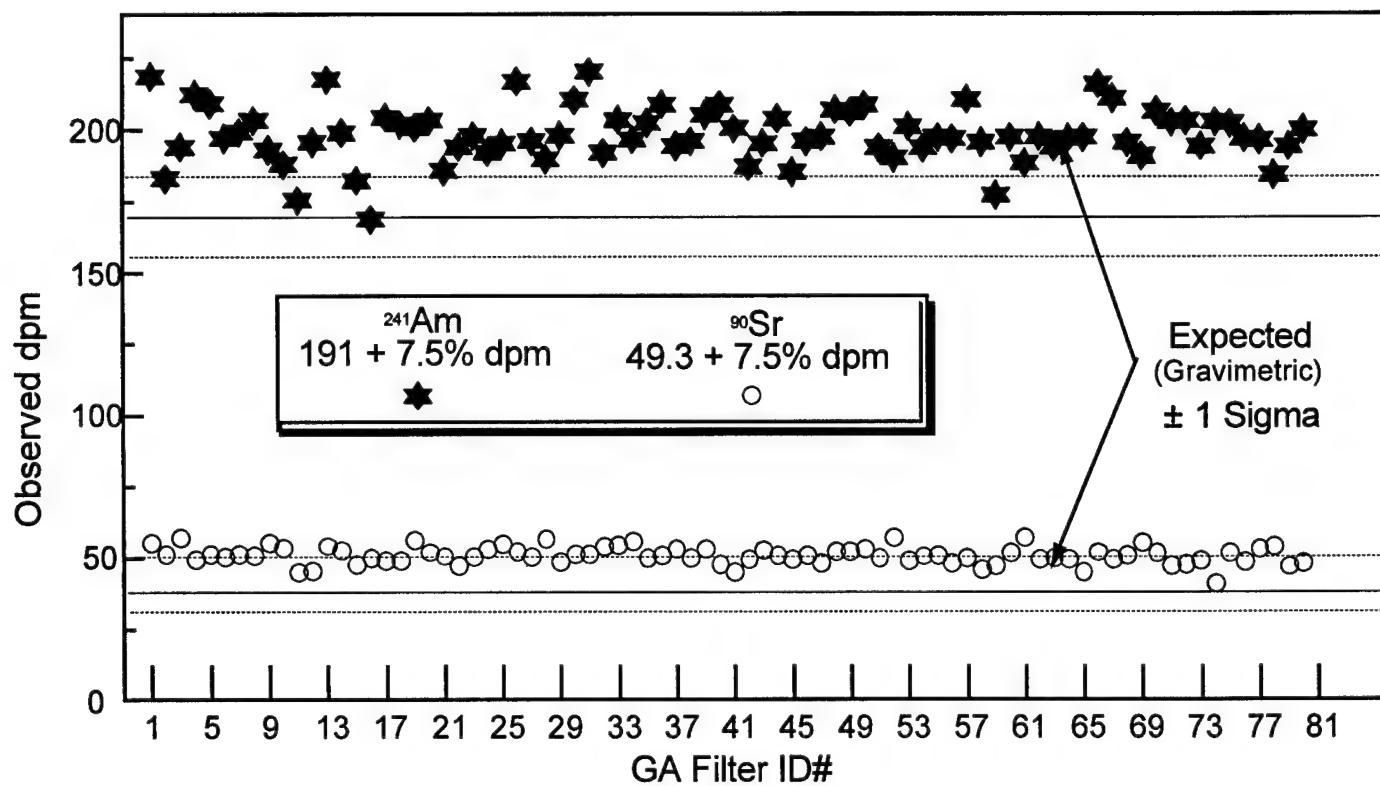


Figure 9. Data summary of gross alpha air filters using Tennelec LB4100W.

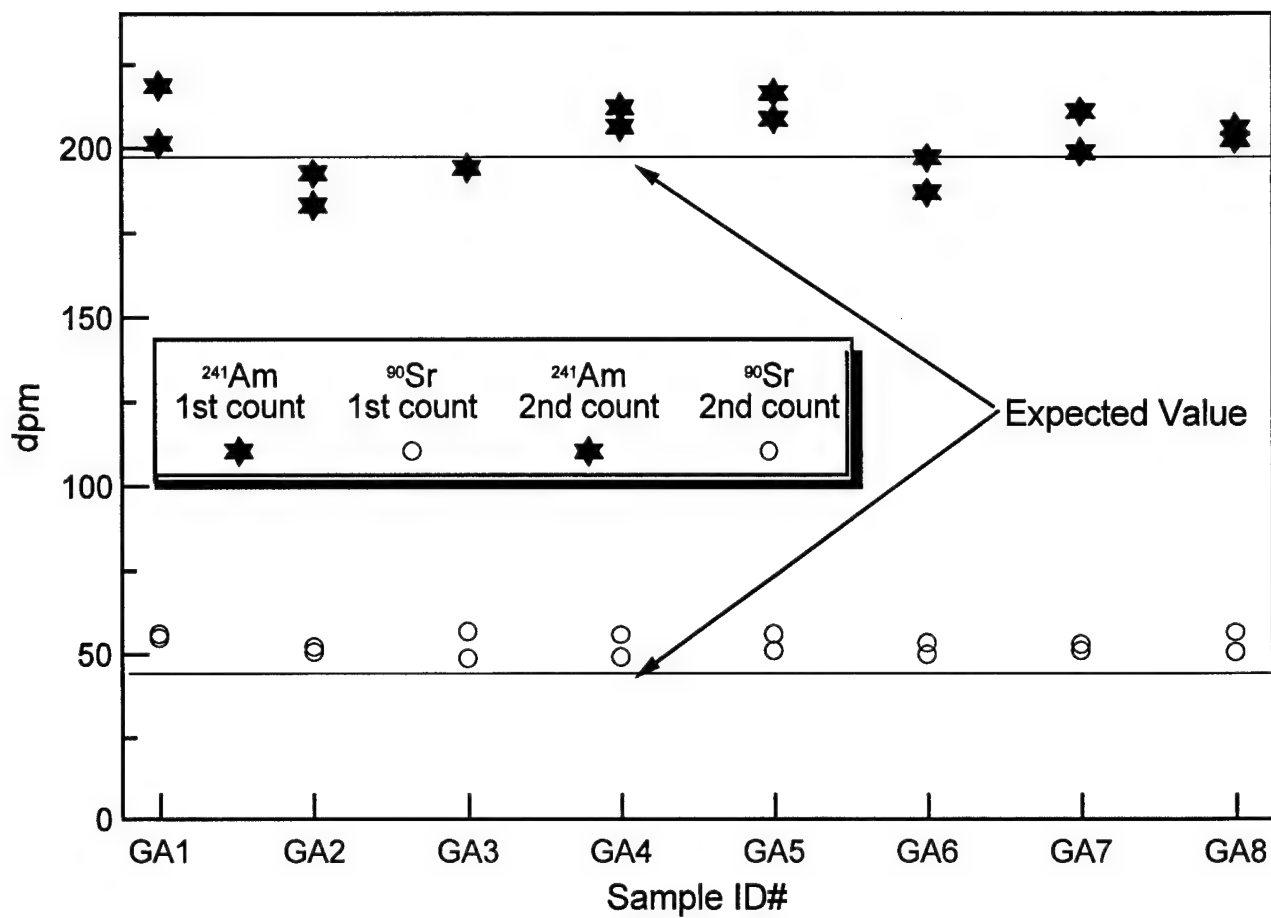


Figure 10. Reproducibility of Tennelec LB4100W System using eight spiked 45-mm foam pads.

A

PPENDIX I

GROSS ALPHA/BETA CALCULATIONS

Calculate the alpha radioactivity using the following equation:

$$\text{Alpha (Bq L}^{-1}\text{)} = [A \times 1000]/[60 \times C \times V]$$

where:

A = the net alpha count rate (gross alpha count rate minus the background count rate) at the alpha voltage plateau,

C = the alpha efficiency factor, read from the graph of efficiency vs. milligram of solids per cm² of planchet area, (counts min⁻¹ dpm⁻¹),

V = the volume of sample aliquot (mL), and

60 = the conversion factor from dpm to Bq.

Calculate the beta radioactivity using the following equation if there are no significant alpha counts when the sample is counted at the alpha voltage plateau.

$$\text{Beta (Bq L}^{-1}\text{)} = B \times 1000/[60 \times D \times V]$$

where:

B = net beta count rate (gross alpha count rate minus the background count rate at the beta voltage plateau),

D = beta efficiency factor, read from the graph of efficiency vs. milligrams of solids per cm² of planchet area (cpm dpm⁻¹),

When counting beta radioactivity in the presence of alpha radioactivity by gas-flow proportional counting (at the beta plateau), alpha particles are also counted. Because alpha particles are more readily absorbed by increasing sample thickness than beta particles, the alpha/beta count ratios vary with increasing sample thickness. Therefore, preparing a calibration curve by counting standards containing ²⁴¹Am with increasing thickness of solids on the alpha plateau is necessary and then on the beta plateau, plotting the ratios of the two counts vs. density thickness. The alpha amplification factor (E) from that curve is used to correct the

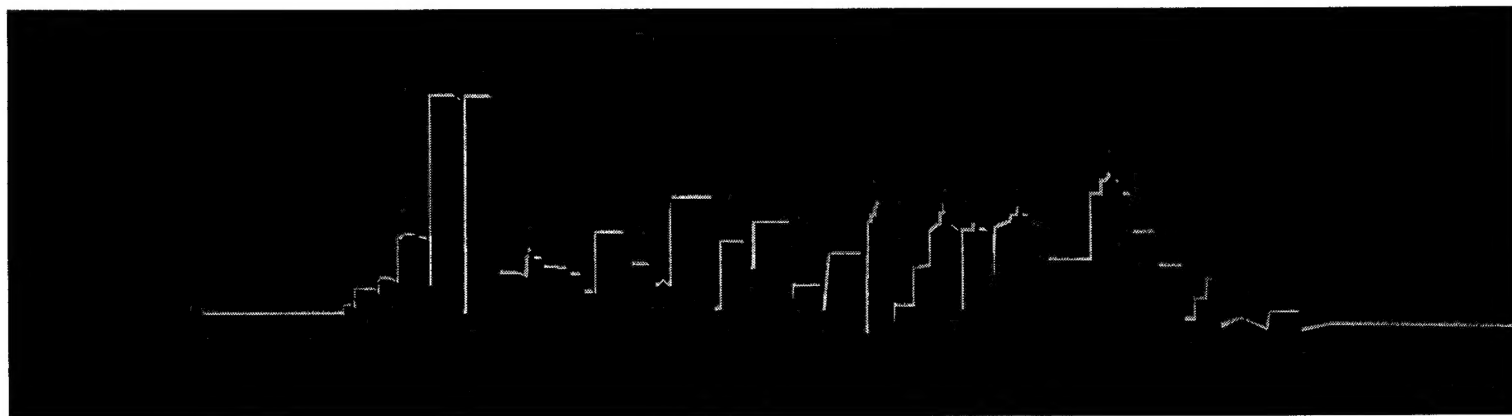
amplified alpha count on the beta plateau. When significant alpha activity is indicated by the sample count at the alpha voltage plateau, the beta activity of the sample can be determined by counting the sample at the beta voltage plateau and by calculating the activity from the following equation:

$$\text{Beta (Bq L}^{-1}\text{)} = (B - AE) \times 1000 / [60 \times D \times V]$$

where:

E = the amplification factor, read graphically from the ratio of alpha counted at the beta voltage:
alpha counted at the α voltage vs. sample thickness.

(Note: These calculations and the errors associated with the results of the analyses are performed by the computer software for gross alpha and beta measurements using gas flow proportional counters.)



EML

Environmental Measurements Laboratory
U. S. Department of Energy
201 Varick Street, 5th floor, New York, NY 10014-4811
<http://www.eml.doe.gov>



INTERNET DOCUMENT INFORMATION FORM

A . Report Title: Preparation and Validation of Gross Alpha/Beta Samples Used in EML's Quality Assessment Program

B. DATE Report Downloaded From the Internet 8/31/98

C. Report's Point of Contact: (Name, Organization, Address, Office Symbol, & Ph #): Environmental Measurements Lab
U.S. Department of Energy
201 Varick Street, 5th Floor
New York, NY 10014-4811

D. Currently Applicable Classification Level: Unclassified

E. Distribution Statement A: Approved for Public Release

F. The foregoing information was compiled and provided by:
DTIC-OCA, Initials: UM **Preparation Date:** 8/31/98

The foregoing information should exactly correspond to the Title, Report Number, and the Date on the accompanying report document. If there are mismatches, or other questions, contact the above OCA Representative for resolution.